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The impact of rainfall in remobilising particulate matter accumulated on leaves of four evergreen species grown on a green screen and a living wall.

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Highlights

- The potential of rainfall to remobilise PM captured on leaves of plants on a living wall and a green screen was studied.
- There was a significant impact of rainfall in washing the PM (PM_1, $PM_{2.5}$ and PM_{10}) off the leaves.
- There was a differential impact of rainfall in remobilising PM on different species of plants.
- A high rainfall intensity had a significant positive impact on PM wash-off from the leaves.

Abstract

Green walls have recently been identified as a green infrastructure (GI) solution to the problem of particulate matter (PM) air pollution. Green wall systems mostly use evergreen plants as the leaves are retained throughout the year; however, researchers have argued that evergreen foliage becomes saturated with PM and fails to capture more due to a long retention time on the leaves. This study evaluated the potential of (simulated) rainfall to remobilise these captured PM and renew the capture ability of the leaf surfaces of four evergreen species (*Heuchera villosa* Michx, *Helleborus* × *sternii* Turrill, *Bergenia cordifolia* (Haw.) Sternb., *Hedera helix L*.) used in a living wall and a green screen located along a busy road in Stoke-on-Trent, UK. The approach used compared PM densities on pre- and post-rain exposed leaf surfaces (using leaf halves of the same leaf) and using a paired t-test to identify any significant reduction in PM due to the rainfall. An Environmental Scanning Electron Microscope (ESEM) and ImageJ image analysis software were employed to quantify the PM densities on leaves. The reduction of PM on leaves, following exposure to 16 $mm.hr^{-1}$ simulated rain in six different rainfall durations was estimated in all four species in order to evaluate any variable impact of rainfall on different species of plants. PM wash-off levels on leaves of *H. helix* by 41 $mm.hr^{-1}$ rain was also evaluated, using the same rainfall durations, to assess any differential impact of rainfall intensity on PM wash-off. This study revealed a significant impact of rainfall in washing the particles off the leaves in all rainfall durations used. A one-way Anova in a General Linear Model showed a differential impact of rainfall in remobilising PM on different species of plants. The rainfall with higher intensity (41 $mm.hr^{-1}$) showed a significantly higher impact on PM wash-off compared to 16 $mm.hr^{-1}$ rain. The results of this study demonstrated the potential of green walls to act as good PM traps throughout the year by recycling their capture surfaces.

Key words: PM wash-off; simulated rain, evergreen species; leaf micromorphology; Green Infrastructure

1. Introduction

Particulate matter (PM) less than 10 µm in aerodynamic diameter (PM_{10}) was graded as the most influential contributor to outdoor air pollution due to the higher prevalence of morbidity caused by PM in comparison to other air pollutants; 90% of urban dwellers were estimated to have been exposed to PM pollution which exceeded WHO standards in 2014 (WHO, 2016). An estimated 428,000 premature deaths were caused by $PM_{2.5}$ (PM less than 2.5 µm aerodynamic diameter) in Europe in 2014 (Guerreiro *et al*., 2017), and 29,000 annual premature deaths in the UK were attributed to PM pollution (RCP, 2016). The International Agency for Research on Cancer categorized PM as a human carcinogen (IARC, 2015) mainly relating to its impact on lung cancer (Pope III, 2002; Raaschou-Nielsen *et al*., 2016); in addition, childhood leukemia and bladder cancers are also known to be associated with PM pollution (Raaschou-Nielsen *et al*., 2017). Short-term and long-term exposure to PM pollution is also a factor in several respiratory diseases such as asthma, cardio-vascular diseases (Anderson *et al*., 2013; Pope III, 2002; Seaton *et al*., 1995), and has been linked to neurodegenerative diseases via particles in the ultrafine range entering the brain (Maher *et al.*, 2016).

Studies have revealed that vegetation has a great potential for removing PM pollutants from the atmosphere (Beckett *et al*., 2000; Blanusa *et al*., 2015; Dover and Phillips, 2015; Freer-Smith *et al*., 2005; Jin *et al*., 2014; McDonald *et al*., 2007; Nowak *et al*., 2013; Song *et al*., 2015; Weerakkody *et al.,* 2017). The use of vegetative barriers to filter PM pollutants, thereby improving human health and wellbeing, is of particular interest, especially when implemented at the city scale (Baldauf, 2017; Lin *et al*., 2016; Tong *et al*., 2016). Irrespective of the type of Green Infrastructure (GI) used or the study location, many reports have demonstrated that some species of plant are better than others in capturing PM (Currie and Bass, 2008; Freer-Smith *et al*., 2005; Leonard *et al*., 2016; Liang *et al*., 2017; Sæbø *et al*., 2012; Song *et al*., 2015; Weerakkody et al., 2017) thus emphasising the need for careful species selection. Evergreen species can be particularly useful in countries where there is seasonal variation, as their foliage is retained throughout the year, and their high PM removal efficiency has been repeatedly reported in previous studies (Beckett *et al.*, 2000; Dochinger, 1980; Hwang *et al*., 2011; Wang *et al.* 2011). However, in a comparison of evergreen conifers and deciduous species, Beckett *et al*. (2000) stated that leaf needles of conifers are retained for several years without being shed and become saturated with particles. In addition, they have the potential to accumulate toxic chemicals over their extended lifespan whereas deciduous species, with broader leaves, have the advantage that captured particulates can be shed at leaf-fall and new shoots and buds provide new capture surfaces. It has also been argued that the accumulated PM on leaves can be remobilised either by re-suspension to the atmosphere by wind (Currie *et al.,* 2008; McPherson *et al.,* 1994) washed off by rain (McPherson, *et al*., 1994; Przybysz *et al*., 2014; Schaubroeck *et al*., 2014; Wang *et al*., 2015; Xu *et al*., 2017) or concentrated at the tips of the leaves by rainfall (Van Bohemen *et al*., 2008). If this is indeed the case, then the PM capture surfaces of leaves of both evergreen and deciduous species is probably regularly restored without saturation occurring – provided the local environment supports resuspension.

Vertical greenery systems have recently been recognised as a short term GI solution to PM pollution (Cheetham *et al*., 2012; Perini *et al*., 2011; Weerakkody *et al*., 2017). Plant species used in these systems are mostly evergreen as their foliage is retained throughout the year maintaining their aesthetic value. Therefore, understanding the leaf PM wash-off behaviour of plants used in the various types of green wall, in order to be able to select the most appropriate species (i.e. those whose surfaces can continue trapping particles without saturation), is important.

Schaubroeck *et al*. (2014) developed a multilayered PM removal model to study the influence of weather on PM remobilisation in forest canopies of *Pinus sylvestris*; however, applying their findings to urban shrubs, herbs or vertical greenery systems is problematic due to their different configurations. There have been few attempts made to quantify the PM remobilisation ability of rainfall at the leaf level. Wang *et al*. (2015) quantified PM reduction on leaves of an evergreen tree, *Ligustrum lucidum* by collecting its leaves after natural rainfall events; however, rainfall intensities, continuity, and intervals between rainfall and sampling were not specified. Although it is difficult to mimic a natural rainfall event to meet all potential conditions, rain simulation can mimic certain important attributes of rain to provide information on PM reduction under specific rainfall intensities and volumes. Perini *et al*. (2017) evaluated PM reduction due to rainfall by washing leaves in water, an approach which might not provide an accurate simulation of rainfall as washing in water cannot simulate specific/different rainfall intensities, amounts of rain, or the kinetic energy carried by raindrops (Neinhuis and Barthlott, 1998; Wang *et al*., 2015; Xu *et al*., 2017). Przybysz *et al*. (2014) attempted to provide a similar effect to rainfall by spraying 20 mm of water on leaf needles of *Pinus sylvestris* (Scots pine), though there was no information on rain intensity, duration or continuity. Xu *et al*. (2017) carried out a comprehensive study on PM wash-off on leaves of three deciduous species of trees and one evergreen shrub by simulating three rainfall intensities using an artificial simulator. In this experiment, plant twigs were exposed to simulated rainfall, the washed-off PM was collected, and the amount of PM in the wash-off was then estimated gravimetrically; unfortunately, they did not report information on the size ranges of PM washed-off/retained by leaves.

The aim of this study was to evaluate the impact of rainfall in remobilising PM accumulated on leaves of some evergreen species used in two vertical greenery systems: a green screen and a living wall. Green screens are free-standing green facades that use climbing or hanging species rooted in the ground which grow upward by twisting around a supportive structure (e.g. wire mesh) (Chiquet, 2014; Dover, 2015). Living walls are vertically grown, artificially irrigated, mostly hydroponic systems, which facilitate the growth of a wide variety of plant species, with a potential for greater artistic expression than simply using climbing species (Dover, 2015). In both these systems, the leaf configuration is similar, both have a more vertical leaf orientation (i.e. tips toward the ground) than plants grown at ground level, which possibly enhances PM wash-off. Our rainfall simulation approach was similar to the approach of Xu *et al*. (2017). However, in contrast to the gravimetric approach towards evaluation of PM wash-off used by Xu *et al*., (2017), we used Particle Number Concentration (PNC) to ensure that the water soluble fraction of PM, which accounts for about 45% of particulates (Li *et al*., 2012), was included in the analysis. Wash-off levels were estimated with reference to the main PM size fractions discussed above (i.e. PM_{10}, $PM_{2.5}$ and PM_{1}) rather than to total suspended PM, due to their differential deposition rates/aerodynamic behaviour and health effects (Davidson and Wu, 1990; Slinn, 1982). The overall aim was to develop a greater understanding of the effectiveness and suitability of using vertical greenery systems in PM reduction.

2. Material and methods

2.1 Study site and species description

The study was conducted in a laboratory on the campus of Staffordshire University, Stoke-on-Trent, UK using artificial rain simulators (see Section 2.3). The leaves of four plant species, three from a living wall system (Nemec Cascade Garden Ltd.), *Heuchera villosa* Michx. (hairy alumroot), *Helleborus* × *sternii* Turrill (blackthorn strain) and *Bergenia cordifolia* (Haw.) Sternb. (Elephant's ears), and one from a green screen (Mobilane ®), *Hedera helix L*. (English ivy), both located on the University campus, were used in this study (Fig. 1). All four species are evergreen and are commonly found in vertical greenery systems in Europe. Plants were selected to represent different leaf morphological types: *H. villosa* with hairy and velvety leaves of *H. sternii* with ridged and slightly waxy leaves, *B. cordifolia* with smooth and glossy leaves, and *H. helix* with leaves with thick epicuticular wax. The closest potential PM source for both the systems was road-traffic due to their close proximity (5 m) to a busy ‘A’ road, Leek Road with a 20,251 average daily traffic flow (Department for Transport, 2017). As both the systems were located at the same site, both facing the road, and both the same distance from the roadside, all four species were exposed to similar environmental and pollution conditions.

2.2 General experimental approach

The experimental approach used was to estimate of PM wash-off by rainfall by comparing the PM held on two equal halves of leaves exposed to roadside pollution (Ottelé et al., 2010): one half was evaluated immediately after removal from the plant, and the other evaluated after exposure to simulated rainfall. A rainfall event simulating 16 $mm.hr^{-1}$ rain was used to evaluate the impact of rainfall on PM remobilization on leaves of all four species in six different rainfall durations; 16 $mm.hr^{-1}$ is within the normal range of rainfall experienced in the UK (Data.Gov.UK^{Beta}). As leaves of *H. helix* were predicted to show very low PM wash-off rates (Ottelé et al., 2010), due to their waxy epicuticles retaining impacted particles, *H. helix* was selected to evaluate any differential impact of an intense rainfall event of 41 $mm.hr^{-1}$ in six different rainfall durations.

2.3 Design and operation of rainfall simulators

An Environmental Chamber (FMH1/900/-40/+150/P/R, JTS Ltd.) with 1.1 m wide x 1.5 m high x 1.2 m deep internal dimensions was used to simulate 41 $mm.hr^{-1}$ rainfall (Fig. 2a). The environmental chamber was equipped with four plastic spray nozzles, a humidity control fan, and a flow meter. The rain profile required was programmed and operated with a timer for specific rainfall conditions and durations. The minimum rainfall intensity that could be achieved with a consistent flow rate (41 $mm.hr^{-1}$), the mean ambient temperature in Stoke-on-Trent during the period of experimentation (17°C), and six rainfall durations (10, 20, 30, 40, 50 and 60 minutes) were used to run the rain profile.

As the Environmental Chamber could not simulate more typical rainfall intensities with consistent flow rates, a handmade rain simulator (Fig. 2b) was designed to simulate an average rainfall event. The simulator was set up in the laboratory using a commercially available watering kit containing small plastic spray nozzles, a Poly-ethylene (PE) hose with 4 mm diameter and 5 m length, and a quick-fit hose socket (hosepipe connector with a regulator) with 2 l hr^{-1} maximum flow rate (Yanqh48 Pvt. Ltd.). Three spray nozzles were connected to the PE hose and they were held at 1.5 m height above the leaf holder (Fig. 2b). The system was connected to the mains water supply using the quick-fit hose socket. Calibrated glass beakers were arranged on a deep plastic tray completely covering the basal area of the simulator and the simulator was optimised to simulate a 16 $mm.hr^{-1}$ rainfall by adjusting the regulator.

2.4 Leaf sample collection

Sample collection and storage in this study followed the same approach given in Weerakkody *et al*. (2017). Sampling was carried out in daylight under non-rainy weather conditions with at least three non-rainy days prior to sampling. In order to explore the effect of heavy rainfall (41 $mm.hr^{-1}$) on PM remobilization, 20 leaves of *H. helix* were removed from green screens on six separate days (120 leaves in total) during the period March-April 2016. To explore the impact of more typical rainfall levels (16 $mm.hr^{-1}$) on PM remobilisation, 120 leaves per species from all four species of plants were sampled on 12 separate days in March – April 2017. All the species were equally sampled on each occasion (ten leaves/day) to avoid any differential influence of weather on existing PM levels between sample days. Samples were arranged in plastic storage boxes and sealed with minimal disturbance and transferred to the laboratory.

2.5 Simulating the effect of rainfall on leaves

In both the experiments, all the leaves were sectioned into halves at the midrib, one half with the petiole attached and the other without. The latter half of each leaf was used to quantify existing PM levels using the ESEM/ImageJ approach (Section 2.6). The remaining halves were attached to a 1 m-long plastic ruler using adhesive tape via their petioles. In the first experiment, with higher rainfall intensity, the ruler holding the leaves was placed inside the environment chamber at 50 cm height from its floor (Fig. 2a) with tips pointing down at a slight angle. The chamber door was locked and the rain profile was run for a particular time period on each sample day, i.e. a different time duration on each day. Time durations were 10, 20, 30, 40, 50 and 60 minutes producing 6.8 mm, 13.6 mm, 20.5 mm, 27.3 mm, 34.2 mm and 41.0 mm rainfall amounts. Once the rain stopped, leaves were carefully removed and stored in clean plastic containers using blue tac to attach them by their petioles to the container walls (avoiding the leaf surfaces touching each other or the container surfaces). The storage containers, with leaves, were kept open (to aid evaporation of rain water) in a closed drying chamber at 25°C for two hours and PM levels retained on the leaf surfaces were then quantified using ESEM/imageJ analysis. In the experiment using 16 mm.hr^{-1} rainfall, leaf halves of four species were attached to a ruler in random order following the same approach as above. The ruler, with leaves, was subsequently placed in the rainfall simulator by attaching it to the side-walls of the tray (Fig.1b) and exposed to rain for the same six durations used in Experiment 1 producing 2.6 mm, 5.33 mm,8 mm, 10.66 mm, 13.33 mm and 16 mm of rainfall. Leaf drying and quantification of the retained PM followed the same approach as above.

2.6 ESEM/ImageJ approach to estimation of PM levels on leaves

The amount of PM on leaves before and after exposure to rain was quantified in terms of PM density (number of PM on a 1 mm^2 section of leaf) using an ESEM and ImageJ image analysis software, following a similar approach to that given in Weerakkody *et al*. (2017). Six leaf sections were excised from each half of leaf blade (avoiding edges, midrib, tip and leaf base) (Weerakkody *et al*., 2017), three sections to image the adaxial surface and three for the abaxial surface. Leaf sections were mounted on small aluminum sample holders using adhesive carbon tabs. Leaf sections were uncoated and scanned under a low vacuum. Three random micrographs per each leaf section were taken at 450x and 1,000x using Back Scattered Electron signals to estimate the mean PM density. Subsequently, micrographs were imported into ImageJ software and the amount of PM in the following size fractions PM_{10} ($PM_{2.5}$ - PM_{10} excluding $PM_{2.5}$), $PM_{2.5}$ (PM_1 - $PM_{2.5}$ excluding PM_1) and PM_1 ($PM_{0.1}$ - PM_1) were quantified using the auto-threshold tool. The mean density of each PM size fraction on both the adaxial and abaxial surfaces of every leaf half (before and after rain) were separately calculated using their respective micrographs (9 micrographs for each half leaf surface).

In order to explain any inter-species variation in PM wash-off associated with leaf surface characteristics, different leaf micromorphological characters (hair/trichomes, grooves and ridges) were scanned using the ESEM at a range of magnifications (100 x, 250 x and 400x) as appropriate. Ten random leaves from each species were imaged to quantify these characters. Visualization of micromorphology followed the same approach given in Weerakkody *et al*. (2018). All the epidermal protrusions were classified as hairs/trichomes and were manually counted and expressed as density of hairs/trichomes per 1 mm^2. In order to estimate leaf roughness, surface grooves and ridges on the micrographs were segmented and classified using Supervised Classification tool in ArcGIS (ArcMap 10.4 © 2015 ESRI) (Weerakkody *et al.*, 2018). Classified features were randomly sampled using 500 random points using the Create Accuracy Assessment Point tool and the area covered by grooves and ridges were estimated as a percentage of the total area. The distribution of leaf epicuticular waxes were observed using the ESEM and recorded.

2.7. Statistical analysis

Any significant reduction in PM densities in a particular rainfall duration was identified by comparing the two halves of each leaf (pre- and post-rain exposure) using a paired t-test following a Shapiro-Wilk normality test (R statistical software version 3.2.5: R Development Core Team, 2016) and the results expressed as percentage reductions (log transformation of data was carried out where required). Any inter-species variation in PM reduction on the adaxial surfaces of the leaves by rainfall was then identified using a one-way Anova in a General Linear Model (GLM) (R Package: MASS). Significant differences in pairwise comparisons of species were identified and clustered using Tukey's pairwise comparison. Tukey's HSD post-hoc test (package: Agricolae) was used for the pairwise comparison of species and to cluster them into groups based on significant differences in wash-off levels. The impact of different rainfall intensities on PM wash-off from the adaxial surfaces of leaves of *H. helix* was evaluated by comparing the PM wash-off percentages from 16 $mm.hr^{-1}$ and 41 $mm.hr^{-1}$ rainfall, using a student's t-test following a Shapiro-Wilk normality test; as residuals met the assumptions of normality, non-transformed data were used in the analysis (Betts *et al.,* 2007; Wilson *et al.,* 2010). Any difference in wash-off between the adaxial and abaxial surfaces of the leaves of *H. helix* from exposure to 41 $mm.hr^{-1}$ was also compared using a student's t-test.

3. Results

3.1 Reduction of PM density on leaves exposed to rain

Both rainfall intensities used in this study removed significant amounts of all particle size fractions from the adaxial surfaces of the leaves of all the species in all rainfall durations (Fig. 3, Fig. 4a and Appendix). On the abaxial surfaces of the leaves of all species, there was no significant reduction in PM density following exposure to 16 $mm.hr^{-1}$ after 60 mins (Appendix) and, in a few instances, there were slightly higher numbers of PM recorded on post-rainfall samples; the analysis was thus not continued for shorter rainfall durations on abaxial surfaces. Nevertheless, 41 $mm.hr^{-1}$ intensity rainfall did show significant reductions in all PM size densities on the abaxial surfaces of the leaves of *H. helix* at all rainfall durations (Fig 4b and Appendix).

The reduction of all PM size densities on the adaxial surfaces of the leaves of *H. helix* following exposure to 41 $mm.hr^{-1}$ rainfall was significantly greater than those reductions due to 16 $mm.hr^{-1}$ rain in all rainfall durations, except for PM_1 wash-off within 30 minutes and PM_{10} wash-off within 10 minutes (Fig. 5). The differences in PM_{10} reduction densities on the adaxial and abaxial surfaces of leaves of *H. helix* exposed to 41 $mm.hr^{-1}$ rainfall was substantially greater than for the smaller sized particles ($PM_{2.5}$ and PM_1) as a result of significantly higher wash-off levels on the adaxial surface in all rainfall durations (in 10 mins: t =2.66 and p=0.01, 20 mins: t=2.07 and p=0.04, 40 mins: t= 2.13 and p=0.04, 50 mins: t=2.33 and p=0.02, 60 min: t=4.26, p<0.001) apart from that lasting for 30 minutes (t=1.11 and p=0.27) (Fig. 4). PM_1 wash-off levels were not significantly different between the two leaf surfaces except for higher wash-off levels on the adaxial surfaces when exposed for 60 minutes (t=2.23 and p=0.03); similarly, $PM_{2.5}$ densities were not significantly different between the two leaf surfaces except for higher wash-off levels on the adaxial surfaces exposed for 20 (t=2.44 and p=0.02) and 60 minutes (t=3.35 and p=0.002).

Of the three particle size fractions examined, PM_{10} had the highest wash-off levels from the leaves of all four species of plants when exposed to 16 mm.hr^{-1} rain in all rainfall durations and PM_1 showed lowest wash-off levels in most of the rainfall durations in all the species of plants (Fig. 3). However, this pattern was not consistent on leaves of *H. helix* exposed to 41 mm.hr^{-1} rainfall; although PM_1 showed the lowest wash-off levels in most of the rainfall durations (apart from the abaxial surface in 60-minute duration), there were several overlaps between PM_{10} and $PM_{2.5}$ (Fig. 4).

3.2 Inter-species variation in PM wash-off on leaves in exposure to 16 mm.hr^{-1} rainfall

Significant inter-species differences in PM_{10} reduction on the adaxial surfaces of leaves were apparent for all rainfall durations with the exception of 30 minutes (Fig. 6 and Table 1). However, $PM_{2.5}$ showed significant inter-species variation only for the longer time periods (for 40 mins: $F=9.75$ and $p< 0.0001$, 50 mins: $F=10.15$ and $p< 0.0001$, 60 mins: $F=32.55$ and $p< 0.0001$) and PM_1 showed significant inter-species variation only for the longest durations (for 50 mins: $F=11.73$ and $p< 0.0001$, 60 mins: $F=27.44$ and $p< 0.0001$) (Fig. 6, Table 1). Despite this, there was some consistency in PM wash-off levels on leaves of different species of plants at longer time durations, as rainfall durations become shorter this consistent pattern disappears (Table 1). At longer rainfall durations (40, 50, and 60 minutes) *B. cordifolia* showed the greatest wash-off levels for all particle size fractions and a reduction of 82.0% of PM_1, 80.4% of $PM_{2.5}$, and 92.5% of PM_{10} was found following exposure to 16 mm of rain (Fig. 6). The lowest wash-off levels of all particle sizes were mostly found on leaves of *H. helix* and a reduction of 44.30% of PM_1, 48.42% of $PM_{2.5}$, and 71.76% of PM_{10} was following exposure to 16 mm of rainfall.

3.3 Observations of leaf micromorphology

Leaves of different species showed a variable distribution of different micromorphological features which are summarised in Table 2. Leaves of *B. cordifolia* showed the smoothest surfaces on both adaxial and abaxial surfaces compared to others. Both adaxial and abaxial surfaces of leaves of *H. villosa* were hairy with a slightly rough surface texture (Table 2). Leaf roughness of the adaxial surface of *H. × sternii* was higher compared to the other species due to higher numbers of grooves, ridges, and localized layers of epicuticular wax (Table 2). Both the adaxial and abaxial surfaces of *H. helix* were densely waxy with thick epicuticular wax layers.

4. Discussion

Significant reductions in PM densities on the adaxial surfaces of leaves exposed to both rainfall intensities showed that rain can remobilise the particulates accumulated on leaves of all four evergreen species studied (Fig. 3), thereby preventing saturation of leaf surfaces and promoting the capture of particles throughout the year/life of the leaves. Przybysz *et al*. (2014), Xu *et al*. (2017) and Wang *et al*. (2015) reported similar findings using different rainfall volumes; in contrast, Perini *et al*. (2017) found that particles between $PM_{2.5}$ and PM_{10} were not washed-off by rain. The differences in findings between these studies can probably be attributed to the different rainfall simulation methods used; washing leaves in water may not have provided enough intensity and kinetic energy, compared to direct rainfall (Neinhuis and Barthlott, 1998), to remove particles from leaf surfaces. The PM wash-off found for the four evergreen species used in this study when exposed to 16 mm of rainfall was higher (Fig. 3) than the PM reduction of 28% and 48% by 10.4 mm and 31.9 mm rainfall (respectively) on leaves of *Ligustrum lucidum* (Wang *et al*., 2015) and 30% to 40% reduction by 20 mm rainfall on leaf-needles of *Pinus sylvestris* (Przybysz *et al*., 2014). However, rainfall intensities used in these studies were not specified. A similar range of 51% to 70% reduction by 15 mm rainfall was reported by Xu *et al*. (2017) on three deciduous species and one evergreen species, however, it was difficult comparing results of these two studies as they are based on reduction of PM masses rather than PM densities. Xu *et.al.* (2017) also reported that PM can only wash-off when the rainfall exceeds the leaf water-storing capacity. However, even the smallest amount of rain simulated, 2.7 mm (10 minutes of 16 $mm.hr^{-1}$ rainfall) in this study removed significant amounts of particles from the leaves (Fig. 3); in vertical greenery systems, the leaf water-storing capacity might have a reduced impact due to the leaves generally pointing towards the ground. Rainwater can probably more easily flow down the slope of such leaves resulting in higher PM removal rates even with modest amounts of rain. However, running experiments using lower rainfall intensities than 16 $mm.hr^{-1}$ (using more advanced rainfall simulators) might be useful to explore this further.

Following exposure to 16 $mm.hr^{-1}$ rainfall over a full 60-minute period, PM densities on the abaxial surfaces of tested leaves did not show any significant wash-off levels, probably as the underside of the leaves were shielded from the rain (Xu *et al*., 2017). However, intense rainfall (41 $mm.hr^{-1}$) carries more kinetic energy

(Xu *et al*., 2017) which can probably disturb leaf orientation sufficiently to cause them to move, twist, and turn and hence receive rainfall on their abaxial surfaces resulting in PM wash-off. In a real-world scenario, the wind state during rainfall might cause a similar effect and is likely to have an impact on PM wash-off from the leaf surfaces. Linking these explanations to the tree literature is problematic as studies have either analysed the total wash-off levels by rainfall (Wang *et al*., 2015) or the abaxial surfaces were excluded from the experiment due to expected lower contact levels (Xu *et al*., 2017).

In agreement with the studies of Przybysz *et al*. (2014) and Wang *et al*. (2015), larger particle sizes showed higher wash-off rates on exposure to 16 mm.hr^{-1} in most of the rainfall durations whilst PM_1 showed relatively low wash-off rates. This suggests that larger particles were more loosely bound to leaf surfaces compared to smaller particles and/or PM_{10} tends to have a higher fraction of water soluble ions than smaller fractions. However, there was not such a clear pattern in intense (41 mm.hr^{-1}) rainfall, with significant amounts of both $PM_{2.5}$ and PM_{10} being remobilised whilst PM_1 was largely retained. Intense rainfall can probably remove substantial amounts of particles irrespective of their size, unless they are strongly bound to epicuticular surface wax, as are the majority of finer particles (Dzierzanowski et al., 2011; Popek et al., 2013; Terzaghi et al., 2013), or entrenched in fine surface features (Weerakkody *et al.,* 2017). Significantly variable PM wash-off levels resulting from two different rainfall intensities (except for PM_1 wash off within 30 minutes and PM_{10} wash off within 10 minutes) on leaves of *H. helix* (Fig. 5) demonstrated that, a higher rainfall intensity has a positive impact on PM removal from the leaves compared to a lower intensity. Xu *et al*. (2017) found a similar positive impact of high rainfall intensities on PM wash-off; nevertheless, this influence became weaker with increased intensity at 50 mm.hr^{-1} indicating a retained fraction of PM even at higher rain intensities. However, higher PM removal rates on *B. cordifolia* (82. 0% of PM_1, 80.4% of $PM_{2.5}$ and 92.5%) compared to other species, at 16 mm rain suggests that PM retention is probably species specific.

Inter-species variation found in PM wash-off from leaves (Fig. 6) can be, at least partly, attributed to their different surface micromorphologies (Table 2). Leaves of the plants on both the living wall and the green screen had similar leaf configuration and were exposed to similar levels of pollution and weather conditions. Therefore, it is unlikely that there was a differential influence on PM wash-off levels from different plant species due the type of VGS used. According to Xu *et al*. (2017), leaf roughness reduces the kinetic energy of rain that falls onto leaf surfaces. The higher levels of remobilisation of PM on the leaves of *B. cordifolia* compared to other species may thus have resulted from the lack of energy-dissipating structures on its surface. Whilst leaf roughness is likely to reduce PM remobilisation, and smooth surfaces promote it, dense epicuticular waxes on the leaves of *H. helix* may have a role to play in preventing wash-off if it strongly binds PM to the leaf surface compared to the species with sparsely arranged wax plates (Dzierzanowski *et al*., 2011; Ottelé et al., 2010; Terzaghi *et al*., 2013) resulting in relatively lower PM wash-off levels. According to Perini *et al*. (2017), PM (2.5-10 µm) captured on leaves of *H. helix* is not washed-off by rain; in contrast, we found significant wash-off levels in all particle sizes on leaves of *H. helix* with all rainfall durations. When exposed to 16 mm of rainfall (60 minutes duration) 44.30% of PM_1, 48.42% of $PM_{2.5}$ and 71.76% of PM_{10} were washed-off from leaves of *H. helix,* suggesting green screens should act as good PM traps throughout the year. Barthlott *et al.* (1998) described 23 types of cuticular wax, which were structurally varied from thin smooth films to crystalloid projections; hence, they probably have a differential impact on leaf roughness and stickiness. Therefore, to illustrate the impact of leaf wax on PM wash-off needs a comprehensive analysis on different types of wax including their structure and composition. Although leaves of *H. sternii* had localised dense epicuticular wax and a rough leaf surface, the hairy leaves of *H. villosa* retained greater levels of PM compared to *H. sternii* in most of the rainfall durations. The more complex microtopography of hairy leaves (Beckett *et al*., 1998) and their potential secretions (Tomaszewski et al., 2014) might have helped retain particles by interfering with the kinetic energy of rainfall resulting in a reduced PM removal rate. All four species used in this study, despite different leaf morphotypes, showed a considerable potential for rainfall-mediated remobilization of all important particle size fractions and for the restoration of capture surfaces. Hence, the use of evergreen species in living walls and green screens are unlikely to be diminished as PM traps through saturation as suggested by Becket et al. (2000). In non-rainy seasons, spraying water on foliage using a sprinkler or watering hose with a moderate pressure could potentially enhance their benefits as PM filters.

Conclusion

A rainfall with 16 mm.hr^{-1} intensity showed a considerable potential for washing-off PM_1, $PM_{2.5}$ and PM_{10} from the leaves of four evergreen species grown in a living wall and a green screen located at the campus of Staffordshire University, UK. However, there were significant inter-species variations in wash-off PM in most of the rainfall durations used, which can probably be attributed to their different micromorphologies. Larger particle sizes showed relatively higher wash-off levels compared to smaller particles. Smooth-leaved *B. cordifolia* had relatively higher PM wash-off levels from their leaves whilst *H. helix*, with a waxy epicuticle, had the lowest wash-off rates. A higher rainfall intensity of 41 mm.hr^{-1} had a positive impact on PM wash-off from leaves of *H. helix,* resulting in significant wash-off levels on both adaxial and abaxial surfaces of its leaves. The results of this study demonstrated the potential of rainfall for restoring PM capture surfaces of evergreen species used in living walls and green screens suggesting that they have the ability to act as PM filters throughout the year without being saturated by pollutants.

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List of figures

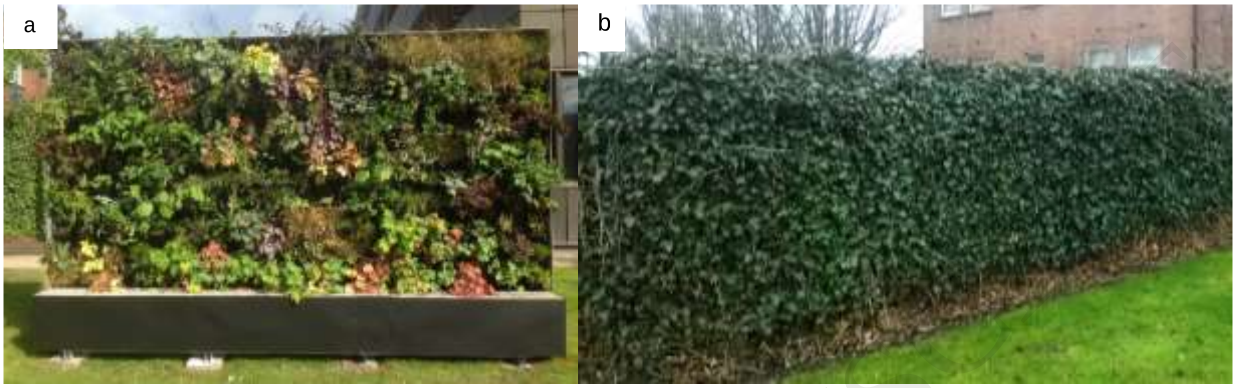

Fig. 1. a) the living wall and b) green screen located on the Staffordshire University campus along Leek Rd, Stoke-on-Trent, UK

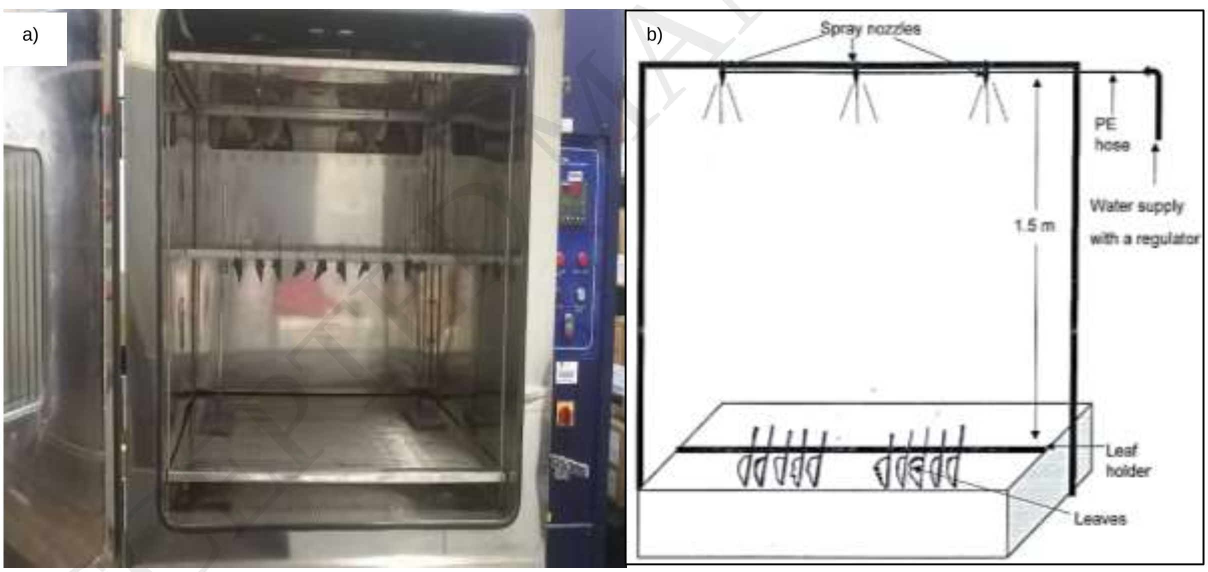

Fig. 2. a) the Environmental Chamber used in the 41 $mm.hr^{-1}$ rainfall study, and b) a schematic diagram of the 16 $mm.hr^{-1}$ simulator used in this study. Note the semi-vertical leaf arrangement in the simulators

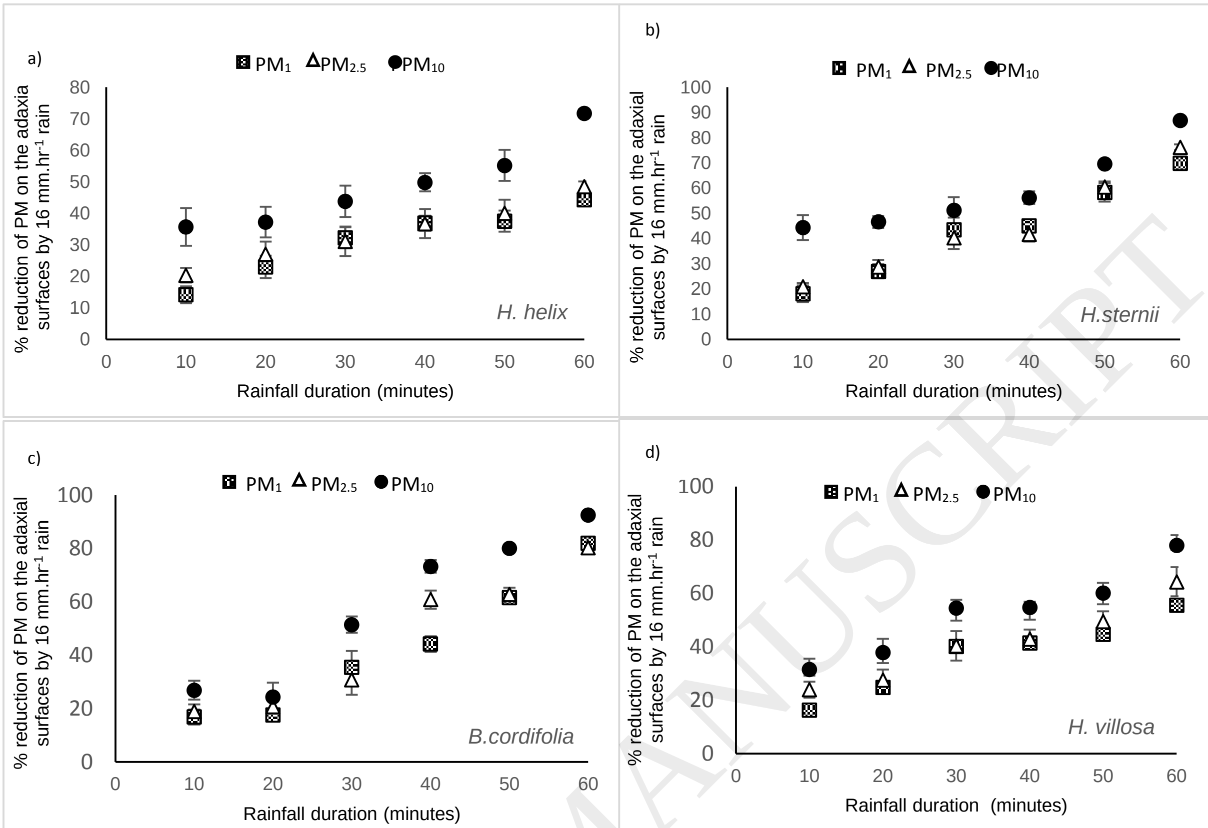

Fig. 3. Reduction of PM densities (%) ± 1SE following exposure to 16 mm.hr^{-1} rainfall on the adaxial surfaces of leaves of a) *H. helix*, b) *H. sernii*, c) *B. cordifolia*, d) *H. villosa.* For statistical comparisons, see the Appendix.

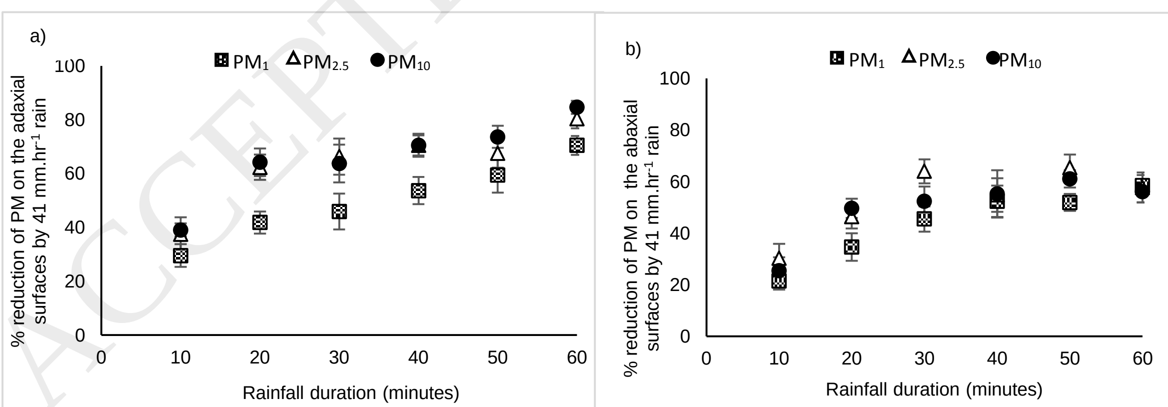

Fig.4. Reduction of PM densities (%) ± 1SE following exposure to 41 mm.hr^{-1} rainfall on a) the adaxial surfaces of *H. helix* and b) the abaxial surfaces of *H. helix.* For statistical comparisons, see the Appendix.

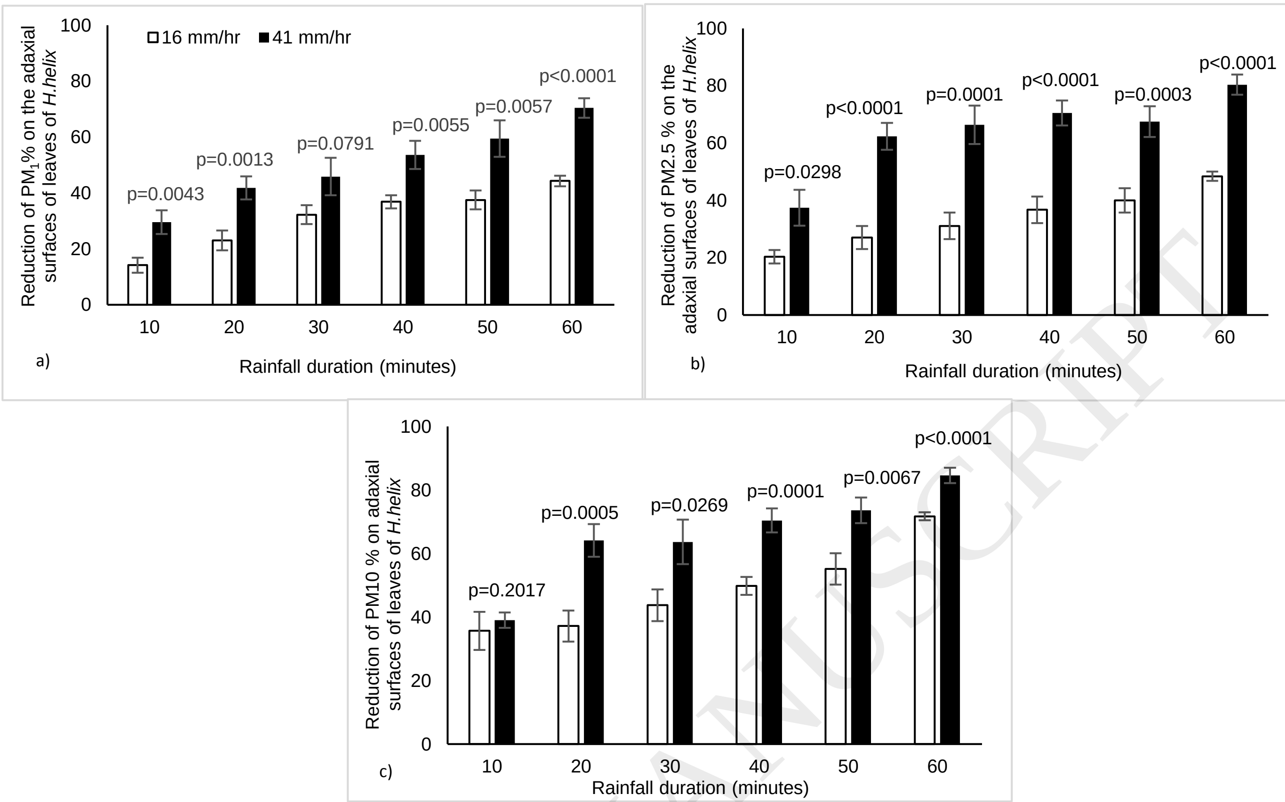

Fig. 5. Reduction of PM densities (%) ± 1SE a) PM_1 b) $PM_{2.5}$ and c) PM_{10} on the adaxial surfaces of leaves of *H. helix* exposed to different durations and intensities of rainfall.

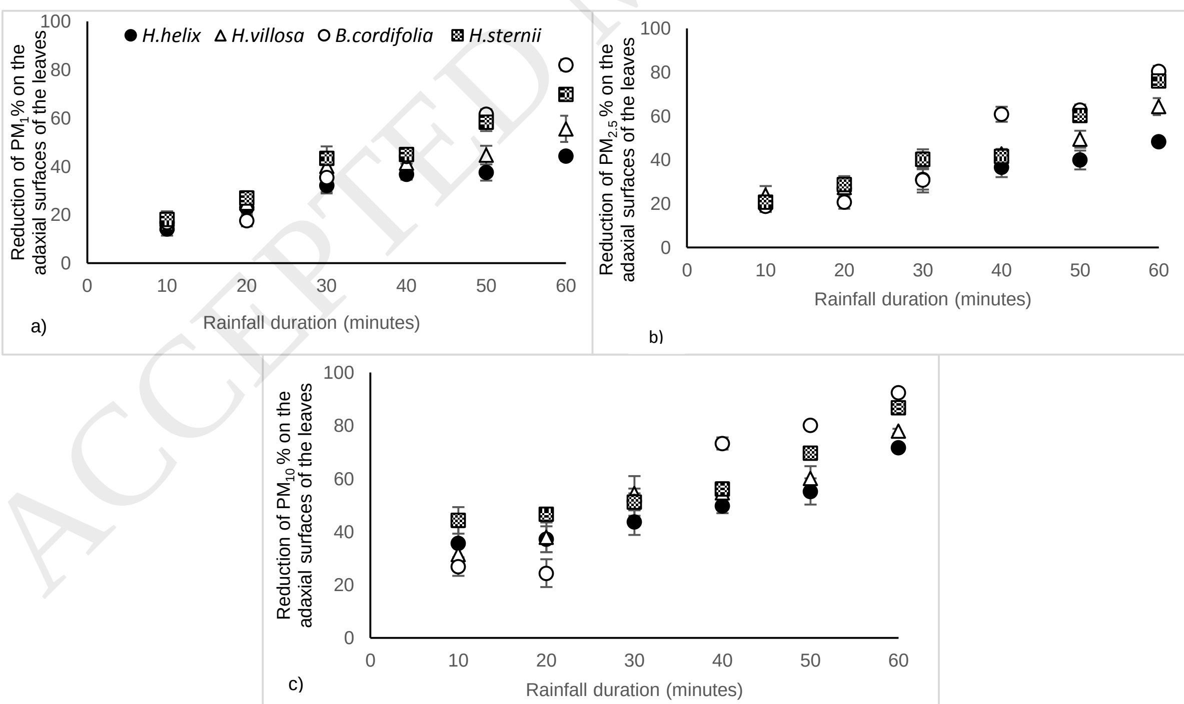

Fig. 6. Reduction of PM densities (%) ± 1SE of a) PM_1 b) $PM_{2.5}$ and c) PM_{10} on the adaxial surfaces of the leaves of different species of plants following exposure to 16 mm.hr^{-1} rainfall of different durations.

Table 1 Results of GLM in the analysis of inter-species variation in PM washed-off from the leaves of different species of plants following exposure to 16 $mm.hr^{-1}$ rainfall of different durations

Duration (min)	PM size fraction	Inter-species variation resulted by one-way Anova	Group assigned by Tukey's HSD test*			
			B. cordifolia	*H. sernii*	*H. villosa*	*H. helix*
10	PM_1	$F=0.31, p=0.81$	a	a	a	a
	$PM_{2.5}$	$F=0.57, p=0.63$	a	a	a	a
	PM_{10}	$F=2.71, p=0.047$	b	a	ab	ab
20	PM_1	$F=1.51, p=0.22$	a	a	a	a
	$PM_{2.5}$	$F=0.87, p=0.46$	a	a	a	a
	PM_{10}	$F=3.76, p=0.01$	b	a	ab	ab
30	PM_1	$F=0.96, p=0.42$	a	a	a	a
	$PM_{2.5}$	$F=1.44, p=0.24$	a	a	a	a
	PM_{10}	$F=0.81, p=0.48$	a	a	a	a
40	PM_1	$F=1.67, p=0.18$	a	a	a	a
	$PM_{2.5}$	$F=9.75, p< 0.0001$	a	b	b	b
	PM_{10}	$F=17.42, p< 0.0001$	a	b	b	b
50	PM_1	$F=11.73, p< 0.0001$	a	a	b	b
	$PM_{2.5}$	$F=10.15, p< 0.0001$	a	ab	bc	c
	PM_{10}	$F=10.3, p< 0.0001$	a	ab	bc	c
60	PM_1	$F=27.44, p< 0.0001$	a	b	c	c
	$PM_{2.5}$	$F=32.55, p< 0.0001$	a	a	b	c
	PM_{10}	$F=74.9, p< 0.0001$	a	b	c	d

Table 2 Mean leaf size ±1SE, LAI ±1SE and mean quantities ±1SE of micro-morphological characters of the leaves of plant species used in the experimental living wall located near Leek Road, Stoke-on-Trent

Species		*B. cordifolia**	*H. × sternii**	*H. villosa**	*H. helix*
Micromorphology on adaxial surface of the leaves ±SE	Density of Hair (mm^{-2})	Not observed	Not observed	58.1 ± 12.52	Not observed
	Grooves%	0.3 ± 0.1	29.6 ± 1.9	21.8 ± 2.5	19.7 ± 1.7
	Ridges %	12.1 ± 0.8	37.4 ± 2.4	15.3 ± 2.0	27.4 ± 1.3
	Leaf wax	smooth and thin	Localised wax layers	Thin and less prominent	Densely waxy surfaces with thick wax layers
Micromorphology on abaxial surface of the leaves ±SE	Density of Hair (mm^{-2})	Not observed	Not observed	56.3 ± 10.51	Not observed
	Grooves%	7.7 ± 1.0	5.9 ± 1.0	16.7 ± 1.4	14.2 ± 0.8
	Ridges %	11.3 ± 1.2	5.3 ± 0.7	25.1 ± 2.7	25.8 ± 1.8
	Leaf wax	smooth and thin	smooth and thin	smooth and thin	Thick wax layers but less dense compared to the adaxial surface.

* data previously given in Weerakkody *et al.* (2018).

Appendix: Results of paired t-tests[a] in the analysis of PM wash-off due to 16mm.hr^{-1} and 41 mm.hr^{-1} of rainfall on the leaves of four evergreen species grown in a living wall and a green screen located on the Staffordshire University campus along Leek Rd, Stoke-on-Trent, UK.

Variation in PM wash-off due to rainfall on the adaxial surfaces of the leaves using a paired t-test															
Duration (min)	*Heuchera villosa* (16 mm.hr^{-1})			*Helleborus* × *sternii* (16 mm.hr^{-1})			*Bergenia cordifolia* (16mm.hr^{-1})			*Hedera helix* (16 mm.hr^{-1})			*Hedera helix* (41 mm.hr^{-1})		
	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}
10	p<0.001	p<0.001	p<0.001	p<0.001	p<0.001	p<0.001	p<0.001	p<0.001	p<0.001	p<0.001	p<0.001	p<0.001	P<0.001	P<0.001	P<0.001
	t = 5.46	t = 5.22	t = 9.35	t = 5.28	t = 12.3	t = 6.39	t = 6.16	t = 6.77	t = 6.75	t = 5.17	t = 7.21	t = 5.93	t = 6.19	t = 3.98	t = 11.71
20	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001
	t = 5.82	t = 4.33	t = 5.69	t = 8.86	t = 8.93	t = 14.8	t = 6.56	t = 5.41	t = 9.25	t = 6.37	t = 6.49	t = 6.54	t = 6.71	t = 4.91	t = 5.41
30	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001
	t = 7.3	t = 9.14	t = 5.41	t = 7.36	t = 7.81	t = 8.14	t = 4.01	t = 5.23	t = 14.8	t = 7.72	t = 5.51	t = 5.8	t = 4.25	t = 5.17	t = 5.17
40	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001
	t = 10.38	t = 13.78	t = 15.35	t = 13.74	t = 9.51	t = 13.91	t = 9.21	t = 11.8	t = 17.53	t = 14.52	t = 6.07	t = 12.16	t = 7.49	t = 9.01	t = 6.14
50	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001
	t = 7.215	t = 8.04	t = 12.13	t = 8.72	t = 19.03	t = 59.4	t = 9.25	t = 9.14	t = 13.1	t = 8.53	t = 8.43	t = 8.21	t = 6.71	t = 8.59	t = 9.32
60	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001	P<0.001
	t = 6.39	t = 12.17	t = 30.93	t = 27.37	t = 32.25	t = 20.04	t = 9.25	t = 9.14	t = 13.05	t = 15.78	t = 16.59	t = 44.42	t = 9.44	t = 8.95	t = 10.44
Variation in PM wash-off due to rainfall on the abaxial surfaces of the leaves using paired t-test															
	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}	PM_1	$PM_{2.5}$	PM_{10}
10	–	–	–	–	–	–	–	–	–	–	–	–	P<0.001	P<0.001	P<0.001
													t = 4.93	t = 7.75	t = 6.71
20	–	–	–	–	–	–	–	–	–	–	–	–	P<0.001	P<0.001	P<0.001
													t = 5.31	t = 5.17	t = 5.17
30	–	–	–	–	–	–	–	–	–	–	–	–	P<0.001	P<0.001	P<0.001
													t = 6.38	t = 7.48	t = 4.98
40	–	–	–	–	–	–	–	–	–	–	–	–	P<0.001	P<0.001	P<0.001
													t = 7.39	t = 2.83	t = 6.30
50	–	–	–	–	–	–	–	–	–	–	–	–	P<0.001	P<0.001	P<0.001
													t = 11.12	t = 7.75	t = 9.61
60	P=0.13	P=0.52	P=0.51	P=0.35	P=0.49	P=0.95	P=0.48	P=0.73	P=0.51	P=0.89	P=0.14	P=0.19	P<0.001	P<0.001	P<0.001
	t = 1.54	t = 0.66	t = 0.66	t = 0.95	t = 0.69	t = 0.05	t = 0.71	t = 0.34	t = 0.66	t = 0.13	t = 1.57	t = 1.36	t = 7.99	t = 6.94	t = 4.49

[a] in all cases n=20 for each species

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